

# Photodetachment study of $\text{He}^-$ quartet resonances below the $\text{He}(n=3)$ thresholds

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The photodetachment cross section of  $\text{He}^-$  has been measured in the photon energy range 2.9...3.3 eV in order to investigate doubly excited states. Measurements were made channel specific by selectively detecting the residual He atoms left in a particular excited state following detachment. Three Feshbach resonances were found in the  $\text{He}(1s2p\ ^3P^o) + e^- (\epsilon p)$  partial cross section: a  ${}^4S$  resonance below the  $\text{He}(1s3s\ ^3S)$  threshold and two  ${}^4P$  resonances below the  $\text{He}(1s3p\ ^3P^o)$  threshold. The measured energies of these doubly excited states are 2.959 260(6) eV, 3.072(7) eV and 3.264 87(4) eV. The corresponding widths are found to be 0.20(2) meV, 50(5) meV and 0.61(5) meV. The measured ener-

gies agree well with recent theoretical predictions for the  $1s3s4s\ ^4S$ ,  $1s3p^2\ ^4P$  and  $1s3p4p\ ^4P$  states, respectively, but the widths deviate noticeably from calculations for  $1s3p^2\ ^4P$  and  $1s3p4p\ ^4P$  states.

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## 1 Introduction

Doubly excited states of two-electron atomic systems provide us with an opportunity to investigate the interplay of electron-electron and electron-core interactions. Negative ions are of particular interest because here the normally dominant Coulomb interactions between the electrons and the core are suppressed, thus enhancing the role of the interelectronic interaction. Pioneering photodetachment experiments on the pure two-electron negative ion  $\text{H}^-$  were first performed by Bryant and co-workers [1]. Several experiments have been performed on  $\text{Li}^-$  [2, 3], which

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can be considered an effective two-electron ion since the two valence electron move in a spherically symmetric potential created by the inert  $\text{Li}^+$  core. The three-electron  $\text{He}^-$  ion is also an effective two-electron system at low photon energies since the 1s core electron remains inert up to energies of 40 eV and only slightly perturbs the motion of the two active outer electrons.

The  $\text{He}^-$  ion is the prototype of an unusual class of negative ions that are not stable, but rather metastable, against decay via autodetachment. The lowest energy state of this ion is the  $1s2s2p\ ^4\text{P}^o$  state, which is bound by 77.516(6) meV relative to the  $1s2s\ ^3\text{S}$  state of He [4]. This state can autodetach into the ground  $1s^2\ ^1\text{S}$  state of the He atom via the relatively weak magnetic interactions. The  $J=5/2$  level, for example, has a measured lifetime of  $350(15)\ \mu\text{s}$  [5], making the  $\text{He}^-$  ion sufficiently long lived to survive a transit through a typical ion beam apparatus.

There have been several previous experimental investigations of the photodetachment of  $\text{He}^-$  [6–11] at photon energies in the visible. Calculations of the  $\text{He}^-$  photodetachment cross section and resonance parameters have also been made [12–20]. In this paper we present the results of a recent photodetachment study of double excitation in the  $\text{He}^-$  ion in the energy range  $2.9\dots3.3\ \text{eV}$ . We have observed one resonance below the  $\text{He}(1s3s\ ^3\text{S})$  threshold and two resonances below the  $\text{He}(1s3p\ ^3\text{P}^o)$  threshold. We have identified the resonances with the  $1s3s4s\ ^4\text{S}$ ,  $1s3p^2\ ^4\text{P}$  and  $1s3p4p\ ^4\text{P}$  states by comparing their measured energies with the results of a recent MCHF calculation of Xi and Froese Fischer [18]. The  $1s3s4s\ ^4\text{S}$  state has been studied previously in a different partial channel of photodetachment [11].

Doubly excited states of  $\text{He}^-$  have also

been studied in electron impact experiments on helium targets [21–24]. Their presence is manifested as resonance structure in scattering cross sections. Many resonance states of doublet symmetry have been observed as transient intermediate states in the electron scattering process. Data on quartet states, however, are sparse. Such states appear as resonances in the cross section of photodetachment from the ground quartet state of the  $\text{He}^-$  ion. The energy resolution in photodetachment experiments is typically much higher than in electron scattering experiments. Thus, the energies and widths of quartet states can, in principle, be measured more accurately than the corresponding doublet states. Selection rules on phototexcitation of  $\text{He}^-$  from the  $1s2s2p\ ^4\text{P}^o$  ground state limit, however, the study of doubly excited states to those with  $^4\text{S}$ ,  $^4\text{P}$  and  $^4\text{D}$  symmetry.

## 2 Experiment

### 2.1 Procedure

The present measurements were made using a collinear laser-ion beam apparatus. The experimental method has been previously used in studies of partial photodetachment cross sections for  $\text{He}^-$  and  $\text{Li}^-$  [3, 11, 25]. It consists of several sequential steps. A doubly excited state of a negative ion is first produced by photoexcitation from its ground state. The excited ion then autodetaches leaving the residual atom, in general, in an excited state. By selectively detecting those atoms left in a particular state one can isolate the corresponding decay channel and investigate the partial cross section. The excited residual atoms are selected by use of the method of resonance ionization spec-

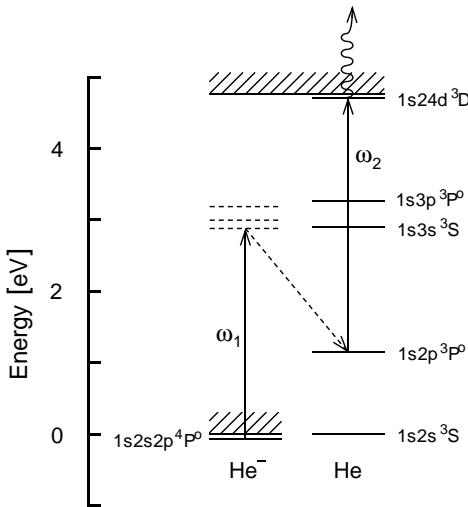
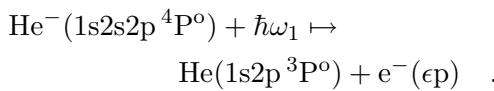


Figure 1: Excitation scheme: selected states of  $\text{He}/\text{He}^-$ . The horizontal dashed lines show the positions of the three measured doubly excited states. The diagonal dashed line indicates auto-detachment of one of these states via the  $\text{He}(1s2p^3P^o)+e^-(ep)$  channel and the wavy line represents electric-field ionization.

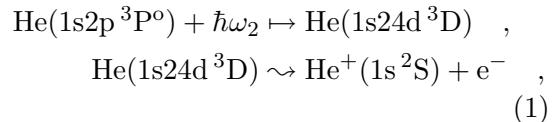
troscopy, i.e. they are further photoexcited to a Rydberg state which is subsequently field ionized in a static electric field. The resulting positive ion signal is proportional to the partial cross section for the selected decay channel.

The investigation of doubly excited states of  $\text{He}^-$  in the present experiment is based on measurements of resonance structure in the partial photodetachment cross section for the  $\text{He}(1s2p^3P^o)+e^-(ep)$  channel. The excitation-detection scheme used is shown in figure 1. In the first step, metastable  $\text{He}^-$  ions are photodetached in a laser field of frequency  $\omega_1$ :



If, in addition, the above process proceeds via an intermediate doubly excited state of  $\text{He}^-$ , resonance structure will appear in the photodetachment cross section. This structure which, in general, has an asymmetric shape, is the result of an interference between the resonant and non-resonant photodetachment processes.

In the second step, He atoms, produced in the  $1s2p^3P^o$  state, as a result of photodetachment, are resonantly excited into the  $24d$  state by a laser field of frequency  $\omega_2$ , sufficiently strong to saturate the transition. These Rydberg atoms are subsequently ionized in a static electric field of about  $200 \frac{\text{kV}}{\text{m}}$ :



where  $\rightsquigarrow$  represents field ionization.

The yield of the  $\text{He}^+$  ions produced in this state-selective detection scheme was recorded as a function of the frequency  $\omega_1$ , while the frequency  $\omega_2$  was held constant on the transition to the Rydberg state. The signal was proportional to the population of helium atoms left in the  $1s2p^3P^o$  state after the photodetachment step. Since the intensity of laser  $\omega_2$  and the ion beam current were constant during a scan, the  $\text{He}^+$  signal, normalized to the intensity of laser  $\omega_1$ , was proportional to the  $\text{He}(1s2p^3P^o)+e^-(ep)$  partial photodetachment cross section. We experimentally determined that this signal changed linearly with respect to the power of laser  $\omega_1$ , thus excluding the possibility of higher order processes.

The detection scheme, based on the selective detection of residual  $\text{He}(1s2p^3P^o)$  atoms, was effective in eliminating a potential background source arising from He

atoms produced in the ion beam via autodetachment of the metastable ground state of  $\text{He}^-$ . These atoms, created in the  $1s^2 1S$  ground state, were not photoionized by laser  $\omega_2$ .

## 2.2 Experimental arrangement

The  $\text{He}^-$  beam was produced from a mass-selected  ${}^4\text{He}^+$  ion beam via charge exchange in a Cs vapor cell. The beam energy was 3.1 keV and the ion current was typically 1 nA.

In the interaction region, shown schematically in figure 2, the laser and ion beams were coaxially superimposed over a distance of 50 cm between the two electric quadrupole deflectors (QD1, QD2). The beam paths were defined by apertures of 3 mm diameter at both ends of the interaction region. The apparatus has previously been described in more detail [26, 27].

One source of background of  $\text{He}^+$  ions arises from double detachment collisions of  $\text{He}^-$  with the residual gas. This contribution was reduced significantly by installing a pair of deflection plates (DP) just before the second quadrupole deflector (QD2). The transverse electric field between the deflection plates was insufficient to field ionize the Rydberg atoms, but strong enough to sweep collisionally-created  $\text{He}^+$  ions out of the beam. To monitor the  $\text{He}^-$  beam current, the deflection plates were periodically grounded.

The highly excited Rydberg atoms of He were field ionized by the static electric field of the second quadrupole deflector (QD2). The resulting  $\text{He}^+$  ions were, in turn, deflected by this quadrupole into the positive ion detector (PD), where they impinged on a metal plate (MP) and produced secondary electrons. These electrons were de-

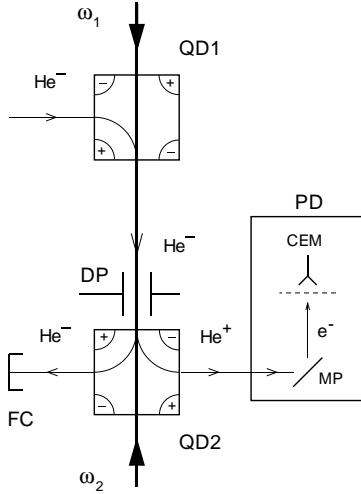


Figure 2: *The interaction and detection regions: QD1, QD2, electrostatic quadrupole deflectors; CEM, channel electron multiplier; DP, deflection plate; PD, positive ion detector; FC, Faraday cup; MP, metal plate.*

tected with a channel electron multiplier (CEM).

The two laser frequencies  $\omega_1$  and  $\omega_2$  used in the experiment were produced by two dye lasers pumped by a common XeCl excimer laser that delivered pulses of about 15 ns duration. The laser light of frequency  $\omega_2$  was generated by PTP dye with a pulse energy of typically 1 mJ. The tunable laser light of frequency  $\omega_1$  was generated by Exalite 416, Exalite 411 and BBQ dyes with pulse energies of typically 1 mJ. The signal was normalized to the power of laser  $\omega_1$ .

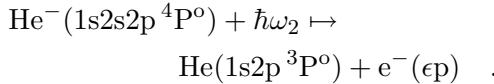
For the measurement of the narrow resonances associated with the  $1s3s4s\ ^4S$  and  $1s3p4p\ ^4P$  states the frequency  $\omega_1$  was calibrated by combining Fabry-Perot fringes with reference lines generated in a hollow cathode lamp. The Fabry-Perot fringes served as frequency markers whereas the reference transitions in argon and ura-

nium provided an absolute calibration of the energy scale. In the measurement of the broad resonance associated with the  $1s3p^2\ ^4P$  state the energy scale was calibrated using our measured position of the  $1s3s4s\ ^4S$  state situated only  $887\text{ cm}^{-1}$  away. In this case a relative scale was established by use of the reading of the laser which has an accuracy of better than  $0.03\text{ cm}^{-1}$  [27].

### 3 Results and discussion

Typical spectral scans of the three resonances in the  $\text{He}(1s2p\ ^3P^o)+e^-(\epsilon p)$  partial cross section are shown in figure 3.

In the case of the  $1s3s4s\ ^4S$  resonance spectrum shown in figure 3(a) there is a small background due mainly to the population of the  $1s2p\ ^3P^o$  state of He by (a) collisional detachment of  $\text{He}^-$  ions and (b) non-resonant photodetachment of  $\text{He}^-$  by laser  $\omega_2$ :



The He atoms formed in the  $1s2p\ ^3P^o$  state by either (a) or (b) will be resonantly ionized according to the processes described in equation (1). The collisional detachment contribution (a) was reduced by maintaining a pressure of  $5\times 10^{-7}\text{ Pa}$  ( $5\times 10^{-9}\text{ mbar}$ ) in the interaction chamber. The contribution (b) was reduced by strongly attenuating the output of laser  $\omega_2$ . The output remained sufficiently strong, however, to saturate the Rydberg transition.

A third source of background (c) can, in principle, make a contribution. The non-resonant photodetachment of  $\text{He}^-$  via the  $\text{He}(1s3s\ ^3S)+e^-(\epsilon s,d)$  channels will populate the  $1s3s\ ^3S$  state of He. This state

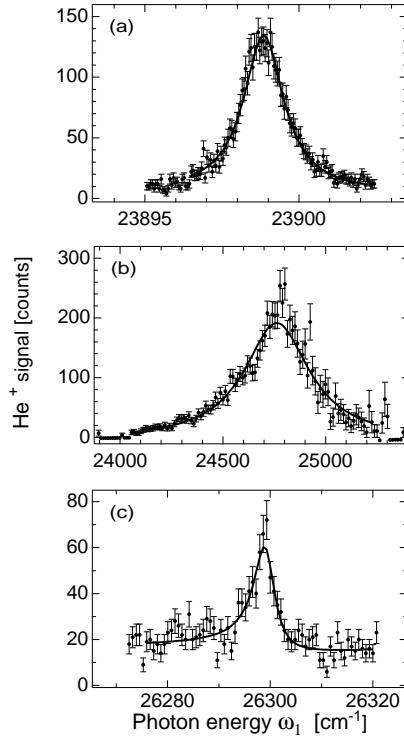


Figure 3: Yield of  $\text{He}^+$  ions vs. photon energy  $\omega_1$ . The solid line is a fit to the experimental data (dots) using equation (2). The three curves show the (a)  $1s3s4s\ ^4S$ ; (b)  $1s3p^2\ ^4P$  and (c)  $1s3p4p\ ^4P$  resonances. The figures show the measured data without corrections for the Doppler shift (see text). Note that the energy scale in (a), (b) and (c) is different.

will subsequently radiatively decay to the  $1s2p\ ^3P^o$  state with a lifetime of 36 ns [28]. This process was found to be negligible when induced by laser  $\omega_2$  due to the previously mentioned attenuation of the output of this laser. It can, however, make a contribution when induced by laser  $\omega_1$ . An exception is the  $1s3s4s\ ^4S$  spectrum where the photon energy  $\omega_1$  is insufficient to populate the  $1s3s\ ^3S$  state of the He atom. The relatively short lifetime of the  $1s3s\ ^3S$  state makes (c) an efficient process for populating the  $1s2p\ ^3P^o$  state. Unfortunately, this contribution is proportional to the intensity of laser  $\omega_1$ , and attenuation of this intensity cannot be used to improve the signal-to background ratio. It, however, remains constant across the scan region of the spectra shown in figure 3(b) and figure 3(c). The signal-to-background ratio is lower for the  $1s3p4p\ ^4P$  spectrum because the peak photodetachment cross section for this resonance is smaller than for the  $1s3p\ ^2\ ^4P$  resonance [29].

The cross section  $\sigma(E)$  in the vicinity of a resonance can be parametrized as [30]:

$$\begin{aligned}\sigma(E) &= a + \frac{b\epsilon + c}{1 + \epsilon^2} \\ \epsilon &= \frac{E - E_0}{(\Gamma/2)} ,\end{aligned}\quad (2)$$

where  $E_0$  is the resonance energy,  $\Gamma$  is the resonance width,  $E$  is the photon energy,  $a$  is the background cross section, and  $b, c$  are the Shore parameters. This function is least-square fitted to the weighted signal and shown as a solid line in figure 3. More than 20 measured spectra were analyzed for each resonance. The background was assumed to be constant over the scan region for all the resonance spectra.

In the case of the narrow  $1s3s4s\ ^4S$  and  $1s3p4p\ ^4P$  resonances data were recorded with both co- and counter-propagating

laser  $\omega_1$  and ion beams. The geometric mean of the measured blue- and red-shifted resonance energies  $E_0^{b,r}$  yields the Doppler-free resonance energy  $E_0 = \sqrt{E_0^b E_0^r}$ . (The energy of the  $^4S$  resonance was found to be

$$E_0^{4s} = 23\,868.031(42) \text{ cm}^{-1}$$

including our previous measurements [11], and the  $1s3p4p\ ^4P$  resonance energy was determined to be

$$E_0^{4p} = 26\,332.97(40) \text{ cm}^{-1} .$$

)<sup>1</sup> For the relatively broad  $1s3p\ ^2\ ^4P$  resonance, it was sufficient to record data with counter-propagating laser  $\omega_1$  and ion beams only. The previously measured  $1s3s4s\ ^4S$  resonance was, in this case, used to calibrate the laser wavelength scale and to account for the Doppler shift.

All measured resonance energies and widths are also given in table 1, together with recent theoretical predictions. In addition, the experimental values of Klinkmüller *et al* [11] measured in another partial channel of photodetachment are given. The quoted uncertainties include both the energy calibration uncertainty and the statistical scatter of the fitted resonance parameters.

The table shows that there is a good agreement between the measured and calculated resonance energies. However there is a noticeable difference between the measured and calculated widths. All four calculations are in essential agreement and all predict the  $1s3p\ ^2\ ^4P$  state to be narrower than is measured. In the case of the  $1s3p4p\ ^4P$  state, the two calculations that predict the width disagree with each other and both values are larger than the measured one.

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<sup>1</sup>The text in brackets is *not* part of the printed version of this publication.

Le Dourneuf and Watanabe [14] have calculated both doublet and quartet doubly excited  $\text{He}^-$  resonances below the  $\text{He}(n=3)$  thresholds using an adiabatic hyperspherical formalism for the two active electrons. They found that the  $\text{He}^+(1s)$  core only slightly perturbed the motion of the outer pair of electrons. In fact, the wavefunctions of the pair displayed correlation patterns very similar to those found in  $\text{H}^-$ . It was suggested that this correspondence allows a rovibrational classification of the doubly excited states of  $\text{He}^-$ . Many resonance states were labeled in this manner, including two that could be identified with the  $1s3s4s\ ^4S$  and the  $1s3p^2\ ^4P$  states observed in the present experiment. The measured width of the  $^4S$  state is very small while that of the  $1s3p^2\ ^4P$  is large. This observation confirms the radial correlation label “-” for the  $^4S$  state and “+” for the  $^4P$  state used by Le Dourneuf and Watanabe.

## 4 Conclusion

In the present experiment the photodetachment cross section of  $\text{He}^-$  has been studied in the photon energy range  $2.9 \dots 3.3$  eV. Three Feshbach resonances associated with the  $1s3s4s\ ^4S$ ,  $1s3p^2\ ^4P$  and  $1s3p4p\ ^4P$  doubly excited states have been observed. The measured energy positions of these states agree well with recent theoretical predictions, but the widths deviate noticeably from calculations.

We hope to continue this work at higher levels of excitation. It will be interesting to observe the evolution of states such as  $1snpn'p\ ^4P$  with increasing values of  $n$ ,  $n'$ . Of special interest are the intrashell resonance states  $1snp^2\ ^4P$  since it is believed that the ladder of such states with increas-

ing  $n$  leads to double detachment. The  $1s2p^2\ ^4P$  state appears as a shape resonance [10] but for  $n>2$  the state is bound with respect to the excited atom and a series of Feshbach resonances appear. These states are the lowest lying in a manifold of  $^4P$  states below the excited state thresholds. Bylicki [19] and Themelis *et al* [16,17] have calculated the energies and widths of these intrashell resonance states up to  $n=7$ . Both groups predict a narrowing of the states with increasing  $n$ . Themelis and Nicolaides show that the configurational composition of these  $^4P$  states changes significantly with  $n$ . For example, in the case of the present work, the resonance at 3.072 eV photon energy has been labeled by the dominant configuration, the  $1s3p^2$  configuration. This is a reasonably valid label since, in this case,  $1s3p^2$  configuration amounts to about 79% of the total composition. As a result of increased angular correlation, however, the contribution from the  $1sn^2$  and other configurations grow with  $n$ . For  $n>4$  the  $1sn^2$  configuration no longer dominates and the label becomes invalid. In this manner we can investigate the gradual breakdown of the independent electron model due to the increased role of correlation in the determining the motion of the active pair of electrons.

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of Sciences through its Nobel Institute of Physics and the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences.

Table 1: Experimental and theoretical resonance energies  $E_0$  (in eV) and widths  $\Gamma$  (in meV). The resonance energies were converted from  $\text{cm}^{-1}$  to eV using the recommended factor  $8\,065\,541\,0 \frac{\text{cm}^{-1}}{\text{eV}}$  [31].

	1s3s4s $^4\text{S}$		1s3p $^2$ $^4\text{P}$		1s3p4p $^4\text{P}$	
	$E_0$	$\Gamma$	$E_0$	$\Gamma$	$E_0$	$\Gamma$
<i>Experiment:</i>						
This work	2.959 260(6)	0.20(2)	3.072(7)	50(5)	3.264 87(4)	0.61(5)
Klinkmüller <i>et al</i> (1997) [11]	2.959 255(7)	0.19(3)	—	—	—	—
<i>Theory:</i>						
Bylicki (1997) [19]	—	—	3.074 24	37	3.264 78	2.45
Xi <i>et al</i> (1996) [18]						
length form	2.959 07	0.19	3.074 70	37.37	3.265 54	1.30
velocity from	2.959 08	0.18	3.074 71	37.37	3.265 47	1.31
Themelis <i>et al.</i> (1995) [17]	—	—	3.096 6	34.6	—	—
Davis <i>et al.</i> (1990) [15]	—	—	3.086 8	33	—	—

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